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X-RAY NONLINEAR OPTICAL PROCESSES IN ATOMS USING A SELF-AMPLIFIED SPONTANEOUS EMISSION FREE-ELECTRON LASER

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X-ray free electron lasers (xFEL) will open new avenues to the virtually unexplored territory of non-linear interactions of x rays with matter. Initially xFELs will be based on the principle of self-amplified spontaneous emission (SASE). Each SASE pulse consists of a number of coherent intensity spikes of random amplitude, i.e. the process is chaotic and pulses are irreproducible. The coherence time of SASE xFELs will be a few femtoseconds for a photon energy near 1 keV. The importance of coherence properties of light in non-linear optical processes was theoretically discovered in the early 1960s. In this contribution we will illustrate the impact of field chaoticity on x-ray non-linear optical processes on neon for photon energies around 1 keV and intensities up to 10^{18} W/cm^2. Resonant and non-resonant processes are discussed.

The first process to be addressed is the formation of a double-core hole in neon by photoionization with x rays above 1.25 keV energy. In contrast to the long-wavelength regime, non-linear optical processes in the x-ray regime are characterized in general by sequential single-photon single-electron interactions. Despite this fact, the sequential absorption of multiple x-ray photons depends on the statistical properties of the radiation field. Treating the x rays generated by a SASE FEL as fully chaotic, a quantum-mechanical analysis of inner-shell two-photon absorption is performed. By solving a system of time-dependent rate equations, we demonstrate that double-core hole formation in neon via x-ray two-photon absorption is enhanced by chaotic photon statistics. At an intensity of 10^{16} W/cm^2, the statistical enhancement is about 30%, much smaller than typical values in the optical regime.

The second part of this presentation discusses the resonant Auger effect of atomic neon at the 1s-3p transition (at 867.1 eV). For low X-ray intensity, the excitation process 1s→3p in Neon can be treated perturbatively. The core-hole excited $1s^{-1}$ 3p state is embedded in the continuum and decays via Auger-process on the timescale of approximately 5 fs.

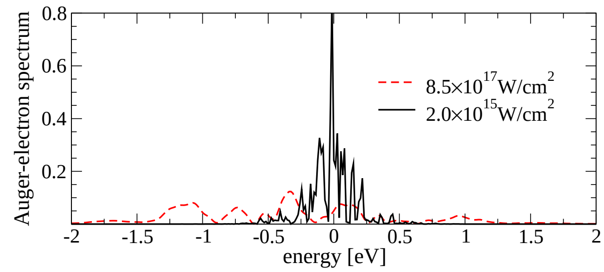

Fig. 1. Resonant Auger-electron line profile for a single shot for two different average intensities. The line profile is extremely spiky, reflecting the multi-mode nature of the SASE pulse. If the x-ray intensity is high enough to induce Rabi oscillations, the electron spectrum gets substantially broader. Pulse parameters: 10^{13} photons per pulse, pulse duration 230 fs, focal diameter 21 μm (solid line), 1 μm (dashed line)

Increasing the x-ray intensity above 1.5×10^{18} W/cm^2, a peak intensity accessible with xFEL sources in the near future, x-ray induced emission from 3p back to 1s becomes possible, i.e. Rabi oscillations between these two levels can be induced. For the numerical analysis of this process, an effective two-level model, including a description of the resonant Auger decay process, is employed. The observation of x-ray-driven atomic populations dynamics in the time domain is challenging for chaotic xFEL pulses. In addition to requiring single-shot measurements, sub-femtosecond temporal resolution would be needed. The Rabi oscillations will, however, be imprinted on the kinetic energy distribution of the resonant Auger electron (see Fig. 1). Measuring the resonant Auger-electron line profile will provide information on both atomic population dynamics and x-ray pulse properties. Prepared by LLNL under Contract DE-AC52-07NA27344.

References

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[2] N. Rohringer and R. Santra, Phys. Rev. A **77**, 053404 (2008).